

QUARTERLY PROGRESS REPORT
ON NASA GRANT NGR 19-005-009

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To

The National Aeronautics and Space Administration

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PURPOSE

The purpose of this report is to give the status of research on NASA Grant NGR 19-005-009. This consist of a summary of all accomplishments to date, and outline which categorize the efforts of the research and the literature survey.

SUMMARY

As shown on the following outline, there are seven (7) major categories for this research. To date items A and B under the heading literature survey, and the design of the reflectometer have been completed. Also, 85% of the equipment have been delivered. However, the monochromator was returned for repairs due to damage caused during shipment.

PURPOSE OF RESEARCH

The purpose of this investigation is to determine original and useful information about the bidirection reflectance of zinc oxide. The bidirectional reflectance will be studied for the spectra between .25-.2.5 microns and the hemisphere above the specimen.

SCOPE OF RESEARCH

This investigation will consider the following factors:

1. Surface conditions
2. Specimen preparation
3. Specimen substrate
4. Polarization
5. Depolarization
6. Wavelength
7. Angles of incident and reflection

The bidirectional reflectance will be checked by experimentally determined angular hemispherical measurements or hemispherical measurements will be used to obtain absolute bidirectional reflectance.

OUTLINE FOR RESEARCH

I. Objective-Scope

II. Literature Survey

- A. Bidirectional Reflectance
 - 1. Experimental
 - 2. Analytical
- B. Angular-Hemispherical
- C. Characterization of Rough Surfaces
- D. Zinc Oxide
 - 1. Physical and Thermal Properties
 - 2. Aging in Ultra Violet and Infrared Light

III. Design of Reflectometer

- A. Sources
- B. Detectors
- C. Monochrometer
- D. Optics
- E. Yoke

IV. Procedure for Obtaining Data

- A. Bidirectional
- B. Angular-Hemispherical
- C. Roughness of Sample

V. Sample Preparation

- A. Repeatability
- B. Coating Technique
- C. Substrates
- D. Oxide Layer

VI. Error Analysis

- A. Variation of Source with Time
- B. Noise
- C. Error in Reflectometer Design

LITERATURE SURVEY

- I. Objectives
- II. Introduction
 - A. Why Study Bidirectional Reflectance
 - B. Scope of Survey
- III. Theoretical Preliminaries
 - A. Radiation Definition
 - B. Electromagnetic Theory
 - C. Predictions from Electromagnetic Theory
- IV. Survey of Experimental Papers on Bidirectional Reflectance
- V. References

OBJECTIVES

- I. To determine the extent of the data available on bidirectional reflectance.
- II. To determine methods of measuring the bidirectional reflectance of zinc oxide.
- III. To determine variables that affect the bidirectional reflectance of zinc oxide.

Introduction

A detailed formulation of radiative heat transfer problems involves the use of the bidirectional reflectance. Except for very simple systems this formulation is very intricate. For this reason and the fact that bidirectional data is scarce, such a formulation is not in common use. However, with the development of the digital computer, there have been numerical methods developed for detailed radiative investigations using the bidirectional reflectance.

Since computations using the bidirectional reflectance are coming into use for spacecrafts radiative studies, it is necessary to have data on the materials involved. Materials that are frequently used for radiative studies are spacecrafts coatings. These coatings are used to aid in controlling the thermal environment of the spacecraft. One of the constituents commonly used for coatings is zinc oxide. This study is primarily concerned with the bidirectional reflectance of zinc oxide.

In preparation for the study of the literature, a review of the electromagnetic theory of the bidirectional reflectance was made. Then a survey of the experimental literature on the bidirectional reflectance was undertaken.

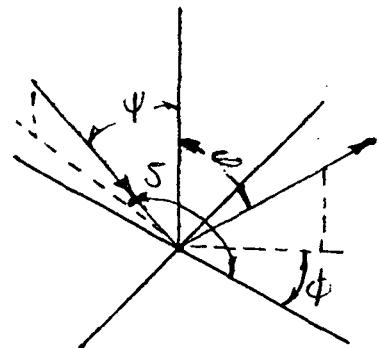
Theoretical Preliminaries

Definition of the Bidirectional Reflectance

The definitions used are those given in the discussion by Torrance and Sparrow at the end of the reference by Birkebak and Eckert (1965).

$$\rho_{ba}(\psi, \varsigma; \theta, \phi) = \frac{dI_r(\psi, \varsigma; \theta, \phi)}{e_i(\psi, \varsigma)}$$

$$= \frac{dI_r(\psi, \varsigma; \theta, \phi)}{i_i(\psi, \varsigma) \cos\psi d\omega_i}$$



$$\rho_{ah}(\psi, \varsigma) = \frac{dC_{r,h}(\psi, \varsigma)}{e_i(\psi, \varsigma)}$$

$$= \int_{\Omega_r} \rho_{ba}(\psi, \varsigma; \theta, \phi) \cos\theta d\omega_r$$

Where

ρ_{ba} is the Bidirectional Reflectance

ρ_{ah} is the Angular Hemispherical Reflectance

e_i is the Incident Energy

dI_r is the Reflected Intensity

i_i is the Incident Intensity

$d\omega_i$ is the Incident Solid Angle

$d\omega_r$ is the Reflected Solid Angle

We also define the relative bidirectional reflectance:

$$\tilde{\rho}_{ba}(\psi, \varsigma; \Theta, \phi) = \frac{\rho_{ba}(\psi, \varsigma; \Theta, \phi)}{\rho_{ba}(\psi_r, \varsigma; \Theta_r, \phi_r)}$$

The reference direction $(\psi_r, \varsigma; \Theta_r, \phi_r)$ is arbitrarily chosen.

The reason for this definition will be given in the discussion of the literature.

ELECTROMAGNETIC THEORY

Introduction

Several theories are used to explain light phenomena. If light interacts with matter with dimensions not of the same order of magnitude as its wavelength, geometrical optics are used to explain the phenomena. If light interacts with matter with dimensions of the same order of magnitude as its wavelength, physical (Electromagnetic Theory) optics are used to explain the phenomena. For interactions of light with atomic entities of matter, quantum optics are used. This division of the theory of light is not precise. Some overlap of the theory does exist.

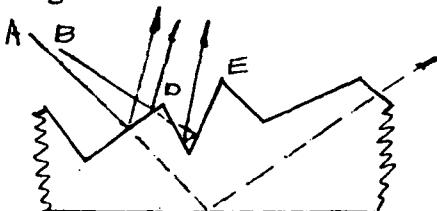
For this study, we are concerned with the Solar Spectrum. The roughness of interest is of the same order of magnitude as the wavelength. Therefore we will be using Electromagnetic Theory to explain the phenomenon of scattering of light from a rough surface. Before a discussion of Electromagnetic Theory and the prediction of the bidirectional reflectance from this theory is undertaken, some factors

which affect the bidirectional reflectance will be discussed.

One of the important phenomena which affects the reflectance is diffraction. Basically this is the bending of light rays when there is an interaction of light with systems with dimensions the same order of magnitude as the light. This phenomenon is used to design diffraction gratings for high resolution monochromators. Electromagnetic Theory (Kirchhoff Method) can be used to predict the nature of the light reflected from such a grating. This phenomenon is the principle reason geometrical optics may lead to erroneous results when used for reflectance predictions.

Phenomena which are difficult to include in analytical predictions are shadowing, multiple scattering and polarization of the light.

The opposite figure shows the multiple scattering of the wave A and B. In addition, it shows the shadowing of facet E by facet D. The multiple scattering is much more unwieldy



for powder samples. This is because the many interfaces cause scattering in all directions.

Light can be considered as being composed of two vectors. These vectors are perpendicular to each other and the direction of propagation. Generally it is found that the reflectance is a function of the particular vector under consideration. As a result, when light is reflected from a surface, the two vectors may not be reflected with the same magnitude. Thus, the reflected light does not have the same polarization as the incident light. This phenomenon is easily studied for smooth surfaces.

Even though this is difficult to analyze for rough surfaces, one should consider it in an experimental investigation.

Another important phenomenon that may exist during reflection investigations is depolarization. Depolarization is the reverse of polarization. For this phenomenon, unpolarized light is observed as the reflected ray for incident polarized light.

MAXWELL'S EQUATIONS

As with any science, Electromagnetic Theory is based upon experimental laws and equations. For a resistor, capacitor and inductor, the governing equations are:

$$I = GV, Q = CV, V = L \left(\frac{dI}{dt} \right),$$

Where

G = Conductance

C = Capacitance

I = Current

L = Inductance

Q = Charge

V = Electromotive Force

When these equations are generalized to the electromotive field, we get for a resistive, capacitive and inductive field:

$$\bar{J} = \sigma \bar{E}, \bar{D} = \epsilon \bar{E}, \bar{B} = \mu \bar{H}$$

The coefficients are related to the electrical circuit quantities,

$$\rho \sim Q, \epsilon \sim C, \mu \sim L.$$

Where

\bar{B} = Magnetic Flux Density

\bar{D} = Electric Flux Density

\bar{E} = Electric Field

\bar{H} = Magnetic Field

\bar{J} = Current Vector

ϵ = Permittivity

μ = Permeability

σ = Conductivity

The governing laws are:

1. Conservation of charge, which leads to the equation of continuity

$$\operatorname{div} \bar{J} = - \frac{\partial \bar{Q}_v}{\partial t}, \bar{Q}_v = \lim_{\Delta t \rightarrow 0} \frac{Q}{\Delta t}$$

2. Ganss' Theorem

$$\iint_S \bar{D} \cdot d\bar{S} = Q.$$

S is a closed surface.

This leads to

$$\operatorname{div} \bar{D} = \bar{Q}_v.$$

3. Amperes Law

$$\oint \bar{H} \cdot d\bar{l} = I$$

This leads to

$$\text{CURL } \bar{H} = \bar{J}.$$

4. Faraday's Law, which leads to

$$-\text{CURL } \bar{E} = \frac{\partial \bar{B}}{\partial t}$$

These laws can be used to obtain the Maxwell Equation. They are:

$$-\text{CURL } \bar{E} = \frac{\partial \bar{B}}{\partial t}, \text{CURL } \bar{H} = \frac{\partial \bar{D}}{\partial t} + \bar{J}$$

$$\text{DIV } \bar{D} = \bar{\rho}_v, \quad \text{DIV } \bar{B} = 0.$$

These equations along with the continuity equation

$$\text{DIV } \bar{J} = -\frac{\partial \bar{\rho}_v}{\partial t},$$

and the constitutive equations

$$\bar{J} = \sigma \bar{E}, \quad \bar{D} = \epsilon \bar{E}, \quad \bar{B} = \mu \bar{H}$$

are used to determine the vector field containing the vectors \bar{B} , \bar{D} , \bar{E} , \bar{H} , \bar{J} , and the scalar $\bar{\rho}_v$. Among the equations above, only six are independent.

The boundary conditions for reflection are:

1. The normal component of \bar{B} at A boundary is continuous;
2. \bar{E} parallel to the surface must be continuous;
3. \bar{H} parallel to the surface must be continuous;
4. \bar{D} normal to the surface must be continuous.

REFLECTION FROM SMOOTH SURFACES

If light is postulated as being an electromagnetic phenomenon, it can be shown from Maxwell's Equations that the mathematical representation of light is a transverse wave. It is sufficient to consider the light vector to be composed of two vectors perpendicular to the direction of propagation and perpendicular to each other. This is the electric field. The magnetic field is perpendicular to the electric field. For analytical studies involving reflection, the light vectors are taken perpendicular and parallel to the plane of incidence. The plane of incidence is the plane containing the incident light ray and the normal to the surface. This is called the P plane. The S plane contains the light ray and is perpendicular to the P plane.

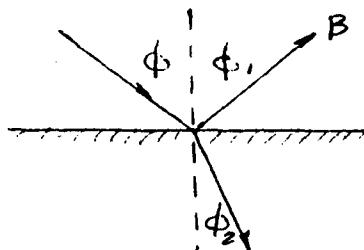
For the interface between two dielectrics, the reflection coefficients are given by the Fresnel Equations,

$$R_P = \left(\frac{E_B}{E_A} \right)_P = \frac{n_2 \cos \phi_i - n_1 \cos \theta_2}{n_2 \cos \phi_i + n_1 \cos \theta_2},$$

$$R_S = \left(\frac{\bar{E}_B}{\bar{E}_A} \right)_S = \frac{n_1 \cos \theta_i - n_2 \cos \theta_2}{n_1 \cos \theta_i + n_2 \cos \theta_2}$$

n_1 and n_2 are the refractive indices of medium 1 and 2 respectively.

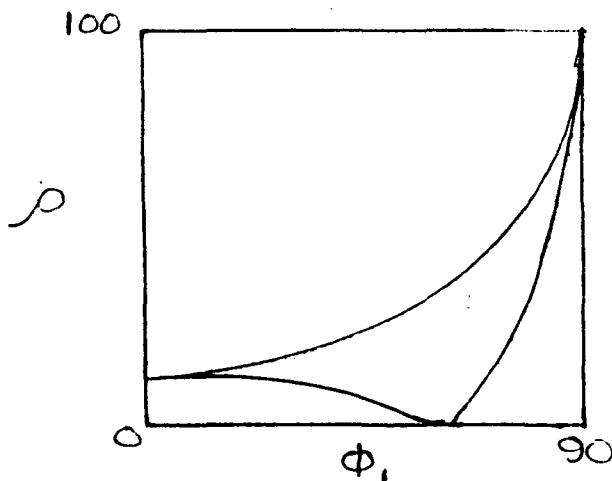
The geometry is shown in the opposite figure.



The power reflectance is given by

$$\rho_s = R_s^2 \quad , \quad \rho_p = R_p^2$$

As shown in the figure below, the E_s and E_p vectors are not reflected equally. This is one of the most important phenomena in light reflection from dielectrics.



REFLECTION FROM ROUGH SURFACES

Many attempts have been made to predict the reflection from rough surfaces. However, the two methods that have had general success are the Kirchhoff and Rayleigh Methods. These methods are described in Beckman and Spizzichino (1963). In the Rayleigh Method, we assume a solution containing an infinite set of plane waves. The boundary conditions and the wave equation are used to determine the amplitudes of these waves. Theoretically the Rayleigh Method looks very good.

However, when we attempt to obtain engineering data with this method, the infinite set of equations that arise to determine the amplitudes prohibit solutions to all but fairly simple problems.

On the other hand, the Kirchhoff Method for a random conducting surface results in a closed form solution. Reflectance predictions using this method are frequently cited in engineering publications on bidirectional reflectance. Some of the details of the formulation of this method will be described below. This description is from Beckman and Spizzichino (1963).

The assumptions or simplifying procedures are:

1. The radius of curvature of the scattering elements is taken to be much greater than the wavelength of the incident radiation
2. Shadowing effects are neglected
3. Only the far field is calculated
4. Multiple scattering is neglected

The wave equation and vector calculus is used to obtain the reflected field in terms of the boundary field. The boundary field is assumed to be given by Fresnel Reflection coefficients. This is the origin of assumption (1) above. For the incident field plane polarized in either the S or P plane, the reflection coefficient for a rough surface is

$$\rho = \frac{E}{E_0} = \frac{1}{4XY\cos\theta} \int_{-X}^X \int_{-Y}^Y (a\zeta'_x + c\zeta'_y - b) e^{i\vec{v} \cdot \vec{r}} dx dy$$

Where

$$\nabla = K \{ (\sin \theta_1 - \sin \theta_2 \cos \theta_3) \bar{x}_0 - \sin \theta_2 \sin \theta_3 \bar{y}_0 \\ - (\cos \theta_1 + \cos \theta_2) \bar{z}_0 \},$$

$$a = (1-R) \sin \theta_1 + (1+R) \sin \theta_2 \cos \theta_3,$$

$$b = (1+R) \cos \theta_2 - (1-R) \cos \theta_1,$$

$$c = (1+R) \sin \theta_2 \sin \theta_3,$$

$$\bar{r} = x \bar{x}_0 + y \bar{y}_0 + f(x, y) \bar{z}_0,$$

$f(x, y)$ is the surface

E is the reflectance from the smooth surface

θ_1 = Incident Polar Angle

θ_2 = Reflected Polar Angle

θ_3 = Reflected Azimuth Angle

x, y = Surface Coordinates

z = Perpendicular to x, y ,

R is not in general the Fresnel coefficients because of local polarization considerations. This is particularly true of dielectrics. Obtaining the correct formulation for R results in unmanageable geometry.

Also in order to obtain a solution, one has to specify the surface

$f(x, y)$. In view of the assumptions on this formulation and the difficulty involved in obtaining a quantitative result, it is clear an experimental investigation is of much more value for engineering purposes.

SURVEY OF PAPERS ON BIDIRECTIONAL REFLECTANCE

It is evident from the analytical discussion that a large number of variables are involved in the investigation of the bidirectional reflectance. This and the fact that a complete determination of the bidirectional reflectance involves many wavelengths and solid angles

has caused most investigators to present minimal data. Data is usually for a restricted number of wavelengths and angles. Many investigations are for the specular plane. A large number are for one incident angle. Still others are for a few and even one wavelength. Generally, published data is not complete enough for detailed engineering calculations.

The intent of most investigators appear to be to give data that indicate characteristics and phenomena. Also, much emphasis is put on correlation of the data. For the most part, attempts are made to correlate data with the results of Davis (1951). Davis obtained a closed form analytical solution for a random conducting surface by employing the Kirchoff Method. In order to use this method for prediction and correlation, it is necessary to experimentally determine two statistical quantities, the standard deviation and a correlation parameter. The parameter that is most often used for correlation is the surface-roughness-to-wavelength ratio.

Torrance (1965) discusses two difficult experimental problems involved in measuring the absolute bidirectional reflectance. The first is due to the limited range of operation of the detectors. Detectors measure energy at a certain level. The band around this level may be several orders of magnitudes. But the difference between the incident energy and reflected energy is a band much larger than the operating range of the detectors. To get around this problem, most investigators present their results as a relative reflectance. This is the reflectance in any direction divided by the reflectance in the specular direction.

In order to measure absolute reflectance, it is necessary to attenuate the incident beam if one detector is used. If two or more detectors are used, it is necessary to calibrate them. Absolute calibration is necessary if their ranges do not overlap. If their ranges overlap, it is only necessary to match their signal-to-energy curves at the overlap point.

The second difficulty is obtaining accuracy when measuring the solid angles. This is best overcome by designing a reflectometer that is simple to machine. Also, to obtain accuracy the design should allow adjustments of critical dimension.

Another measuring factor which is discussed by Love (1907), and Birkebak (1965) is the use of a finite solid angle. The effect of using a finite solid angle is to smooth out any peaks whose width is less than the diameter of the solid angle. For investigations involving materials that have high specular reflection, this is of extreme importance. However, for studies of diffuse samples, the error in data due to the finite solid angle is small.

Table 1 is a summary of bidirectional reflectance data presented in the literature. The first publication to give bidirectional reflectance data is Eckert (1936). As with Munch's (1955) and Middleton's (1952) data, the reflectance is not monochromatic but total. Nevertheless, the reflectance behavior of some important engineering materials are given. The data of Torrance (1965) and many other investigators shows two trends: with decreasing wavelength, the reflectance of a given surface approach that of an ideal diffuse reflector; with increasing wavelength, the reflectance approach those

of an ideal specular reflector. Another trend of importance is discussed by Torrance (1966). This is off specular peaks in the bidirectional reflectance data. Torrance data shows it is possible to have the maximum reflectance in an angle other than the specular angle.

TABLE 1

AUTHOR	WAVELENGTH	ANGLES, DEGREES	MATERIALS
Eckert (1936)	Blackbody	$\Psi \approx 0^\circ$ $\Theta = 0^\circ - 80^\circ$	Various Engineering Surfaces
Munch (1955)	Blackbody 350 - 995 $^{\circ}\text{C}$	$\Psi = 0, 15, 30, 45, 60^\circ$ $\Theta = -90^\circ \rightarrow +90^\circ$ $\phi = 0^\circ$	White typewriter paper Black oxidized brass White pine - irradiated at right angle to fiber direction Colorstred (Stearite substance) Anodically oxidized anticorodal sheet Stanblasted anticorodal sheet
Middleton (1952) Mungall	Visible	$\Psi = 0, 30, 45, 60, 75^\circ$ $\Theta = -80^\circ \rightarrow +80^\circ$ $\phi = 0^\circ$	Snow
Torrance and Sparrow (1965)	.5 - 12 μ	$\Psi = 100 \text{ or } 45^\circ$ $\Theta = 0 \rightarrow 70^\circ$ $\phi = 0, 45, 90, 180^\circ$	Fused Polycrystalline Magnesium Oxide Ceramic
Birkebak and Eckert (1965)	2 - 10 μ	$\Psi = 10^\circ$ $\Theta = 0, 45, 90, 135, 180^\circ$ $\phi = 0 \rightarrow 90^\circ$	Ground Glass Coated with Aluminum, Nickel

TABLE 1 CONT.

AUTHOR	WAVELENGTH	ANGLES, DEGREES	MATERIALS
Torrance and Sparrow (1966)	.5 - 5 μm	$\psi = 10^{\circ}, 20^{\circ}, 30^{\circ}, 45^{\circ}, 60^{\circ}$ $\theta = 0^{\circ} - 50^{\circ}$ $\phi = 0^{\circ}$	Aluminum, Nickel, Copper, Nickel Copper Alloy, Magnesium Oxide Ceramic
Herold and Edwards (1966)	2.5, 5.0, 7.5 μm	$\psi = 10^{\circ}, 20^{\circ}, 30^{\circ}, 45^{\circ}, 60^{\circ}$ $\theta = 0^{\circ} - 50^{\circ}$ $\phi = 0^{\circ} - 180^{\circ}$	Sintered-Bronze, Glass-Beaded Projection Screen, Sand Blasted Aluminum, 100 Mesh Wire-Screen Bonded to mylar, all coated with either Aluminum or Gold
Brandenberg and Neu (1966)	.507, .533 μm	$\psi = 15^{\circ}, 75^{\circ}$ $\theta = 15^{\circ}, 30^{\circ}, 45^{\circ}, 60^{\circ}, 75^{\circ}$ $\phi = 0^{\circ}, 150^{\circ}$	MgO Coating, Barium Sulfate Paint Zinc Oxide Paint, Aluminum
Love and Francis (1967)	.6 - 10 μm	$\psi = 10^{\circ}, 30^{\circ}, 60^{\circ}$ $\theta = 0^{\circ} - 90^{\circ}$	Type 302 Stainless Steel
Smith, Tempelmeyer, Muller and Wood (1969)	.9 μm	$\psi = 10^{\circ}, 30^{\circ}, 50^{\circ}, 70^{\circ}$ $\theta = 0^{\circ} \leq \theta \leq 90^{\circ}$ $\phi = 180^{\circ}, 0^{\circ} \leq \phi \leq 60^{\circ}$	CO_2 Cryodeposits on Polished Copper and Black Epoxy Paint Surfaces
Loehrlein, Winter and Visicanta (1970)	.43, .55 μm	$\psi = 30^{\circ}, 45^{\circ}, 60^{\circ}$ $\theta = 0^{\circ} - 90^{\circ}$ $\phi = 0^{\circ}, 130^{\circ}, 160^{\circ}$	Aluminum, Polycrystalline Magnesium Oxide, Projection Screen, well characterized V-Groove

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